Two-dimensional pump–probe imaging in reacting flows

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We demonstrate that picosecond pump–probe spectroscopy can be used to acquire quantitative two-dimensional images of species in flames, using a loss-modulation-based two-dimensional phase-sensitive detection system mounted in front of a CCD camera. This system is currently limited to detection of 1% modulation depth, but it can be significantly improved. The instrument permits high data collection rates, and it provides a quenching-independent signal without the need for calibration. The signal is embedded in a coherent laser beam, making it possible to perform optical processing before the camera. © 1995 Optical Society of America

Two-dimensional imaging of radical species concentrations in reacting flows (e.g., OH, CH, and NO) has yielded a large amount of useful information for modelers. As one example, planar laser-induced fluorescence (PLIF) has become the technique of choice for turbulent flame studies. Although PLIF has proven invaluable, a number of data manipulations are required to make the measurement quantitative, such as the collisonal quenching correction. In nonpremixed turbulent flames it is difficult to quantify the collisional environment within each pixel area in the flow. This has proven to be a significant limitation to the PLIF technique. There have, in fact, been few quantitative PLIF results reported, even for steady laminar flames.[1]

We recently demonstrated several new diagnostic techniques based on picosecond mode-locked Ti:sapphire lasers.[2,3] The principal advantages of these techniques include: (1) the picosecond pulse diagnostics are not expected to be strongly affected by the collisional environment and (2) the high repetition rates of mode-locked lasers (70–100 MHz) can make it possible to observe all the time scales of importance in turbulence or to observe rapid (submillisecond) transients (e.g., ignition). The pump–probe technique permits the determination of absolute number density, free of corrections and without the need for calibration.[4] The technique therefore has the potential to overcome several of the difficulties associated with PLIF.

In pump–probe spectroscopy the pump beam is modulated, and it is crossed with the probe in an absorbing sample. The pump modulation frequency is impressed on the resonant molecules, which then modulate the probe by means of absorption and stimulated emission. The probe modulation is measured with a photodetector and lock-in amplifier (phase-sensitive detection), and this measurement can be directly related to the number density of absorbers in the overlap between the two beams. We previously seeded K into a CH_4–air flame to demonstrate that the pump–probe technique delivers an absolute K number density at rapid detection rates.[3]

Here we demonstrate that the same technique can be used to acquire two-dimensional images of number density in flames. To perform this experiment, we have expanded the pump beam into a sheet, using cylindrical optics. The probe is then upcollimated into a large-diameter Gaussian-cross-section beam that overlaps the sheet in the flame. It is then necessary to develop a two-dimensional analog to the lock-in amplifier used in the single-point experiments.

In phase-sensitive detection, some input to the experiment (the pump beam here) is modulated (say, at ω_m). That system input produces an output signal (here a modulation on the probe beam) induced by the variable of interest (absorber atoms). The modulated signal has the same frequency as the input, but it can have a phase shift and is typically embedded in a relatively large background consisting of noise components covering a broad range of frequencies (ω_i). In a lock-in amplifier the total signal (modulated signal of interest plus large background) is first demodulated by a mixer. This occurs by multiplication of the total signal by a sinusoid at the chopper frequency:

$$e_i e_m = 0.5 \sum_i (E_i E_m)[\cos(\omega_i - \omega_m)t - \cos(\omega_i + \omega_m)t], \quad (1)$$

producing sum- and difference-frequency terms for all portions of the total signal. The difference term is at zero frequency only for the modulated signal of interest (since ω_i is the same as ω_m in that case). That small portion of the total signal is mixed down to dc because it has the same frequency as the chopper. The output of the mixer is then routed through a low-pass filter, eliminating the sum-frequency term of the modulated signal of interest and any other components (e.g., the large background) that fall outside the bandwidth (user selected) of the filter. The remainder of the signal carries information about the variable of interest (contained in the sideband that passes through the low-pass filter).

Mixers themselves can be thought of as amplifiers in which the amplifier gain is modulated at the chopper

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reference frequency. The amplifier output is the linear product of the input and the gain (sinusoidal), so this performs the multiplication just described. For imaging, it is possible to accomplish the same task by use of optical loss (by polarization modulation) or gain (by use of a microchannel plate or photocathode) directly in front of the camera pixels. If, for example, the reference frequency is used to modulate loss, the optical signal arriving at each pixel is the linear product of the input and the reference waveform. Here we used liquid crystals to perform this task, primarily because they are inexpensive and our goal was simply to demonstrate the concept. It remains to filter this demodulated total optical signal, accomplished here by the integrating behavior of the pixel itself together with the camera framing rate of 30 Hz. This is not necessarily the optimum filter behavior but has proved sufficient for the present demonstration.

One important difference between our two-dimensional optical phase-sensitive detection system and a typical lock-in amplifier is that the demodulator is capable of reducing the light level to zero and then driving it up to some maximum level characteristic of the system (i.e., the optical signal will not become negative). That imposes a dc offset at the electrical input [see Fig. 1(a)], and hence the product of the reference and the signal contains a nonnegligible dc level that is summed with the demodulated signal. It is therefore necessary to subtract this dc component from the total.

Our liquid crystals can modulate light as high as ~200 Hz. For this reason we chose to chop the pump beam and to drive the liquid-crystal demodulator at 30 Hz. The sinusoidal drive for the crystal was synthesized by use of the chopper output. Because of the slow modulation frequency there was effectively no phase difference between the reference and the signal. The demodulation then occurs by means of multiplication of the input by the reference signals [see Fig. 1(a)], resulting in the following expression:

$$e_i e_m = \sum_i \big((\mathrm{dc}_i \mathrm{dc}_m) + [\mathrm{dc}_i E_m \sin(\omega_m t)] + [\mathrm{dc}_m E_i \times \sin(\omega_i t)] + \{E_i E_m [0.5 \cos(\omega_i - \omega_m) \times t - 0.5 \cos(\omega_i + \omega_m) t]\}\big), \quad (2)$$

where the subscript i represents the optical signal and the subscript m represents the liquid crystal. Here $\omega_i = \omega_m = 2\pi \times 30\ (\mathrm{s}^{-1})$ for the signal of interest. For that signal the difference term $[\cos(\omega_i - \omega_m)t]$ goes to 1, and the sum term $[\cos(\omega_i + \omega_m)t]$ is a second harmonic of the 30-Hz signal. The camera framing rate is the same as the chopper rate, 30 Hz, but the signal demodulation and the chopper reference are synchronous. They are not synchronous with the camera, and as such, the camera acts simply as a low-pass filter that does not interact with the mixing process. Such a camera sampling filter curve is shown in Fig. 1(b) (for one camera frame). It has zeros at the framing rate harmonics, which effectively filters all the ω and 2ω terms in Eq. (2). What is left is $(\mathrm{dc}_i \mathrm{dc}_m)$, the dc offset problem, and $(0.5 E_i E_m)$, our signal. A typical lock-in amplifier operated at 30 Hz would require a long time constant to provide good signal-to-noise ratios. That is also the case here, where we integrate over 8–9 s (256 images). To accomplish this, we store a running average on the frame grabber and average each new frame with that image as it is acquired. This is, in effect, exactly what occurs in the filter of a lock-in amplifier. Finally, it remains to subtract the dc offset from the image. To do that, we modulate the crystal at 300 Hz, beyond its ability to modulate light, but preserving the neutral-density value. That eliminates the $[E_i \sin(\omega_i t) E_m \sin(\omega_m t)]$ term but preserves the $(\mathrm{dc}_i \mathrm{dc}_m)$ term. We store the dc image and subtract it from the demodulated image.

For the experiments reported here we used a Perkin-Elmer aspirating burner fitted with a Meeker-type burner head (see Ref. 3 for experimental details). A solution of KCl in deionized water was aspirated into the burner. We used a Spectra-Physics Tsunami Ti:sapphire laser operated at 60 ps and tuned to the $4^2S_{1/2}$–$4^2P^o_{3/2}$ of atomic K at 766.49 nm. The 130-mW pump was spread into a sheet approximately 2.5 cm tall (the flame height) and 1 mm thick. The pump and probe irradiances were smaller than their saturation values.The low-intensity probe was spread into a cylinder ~2.5 cm in diameter and overlapped the pump at 90°. That beam was displayed on a screen, viewed through the liquid crystal by a Javelin CCD camera. The image was acquired and manipulated with a Sharp GPB-1 frame grabber mounted in a personal computer.

Figure 2(a) shows the image of the probe that was visible on the screen while the flame was on and absorbing. Note that the center of the Gaussian probe was off the flame centerline, as shown by the shadow of the burner at the bottom of the photograph. Figure 2(b) shows the demodulated image after the dc term has been subtracted. We were unable to assess the full modulation depth in these experiments owing

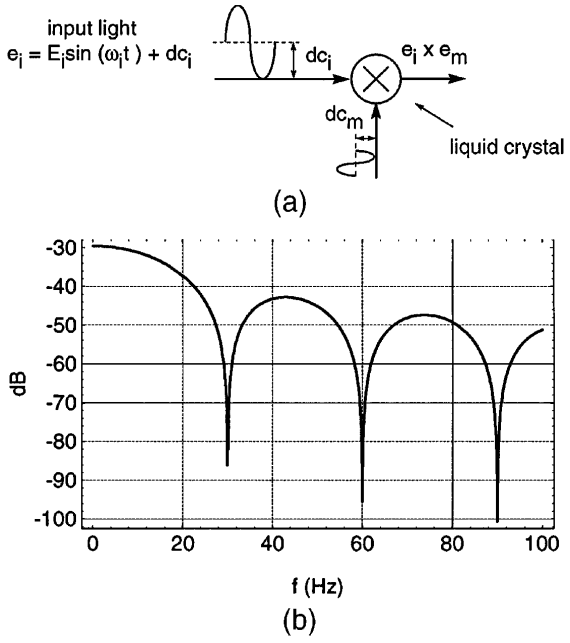

Fig. 1. (a) Schematic of optical demodulation in which the 30-Hz reference signal drives the liquid crystal at $e_m = E_m \sin(\omega_M t) + \mathrm{dc}_m$. (b) 30-Hz sampling filter curve (for one frame).

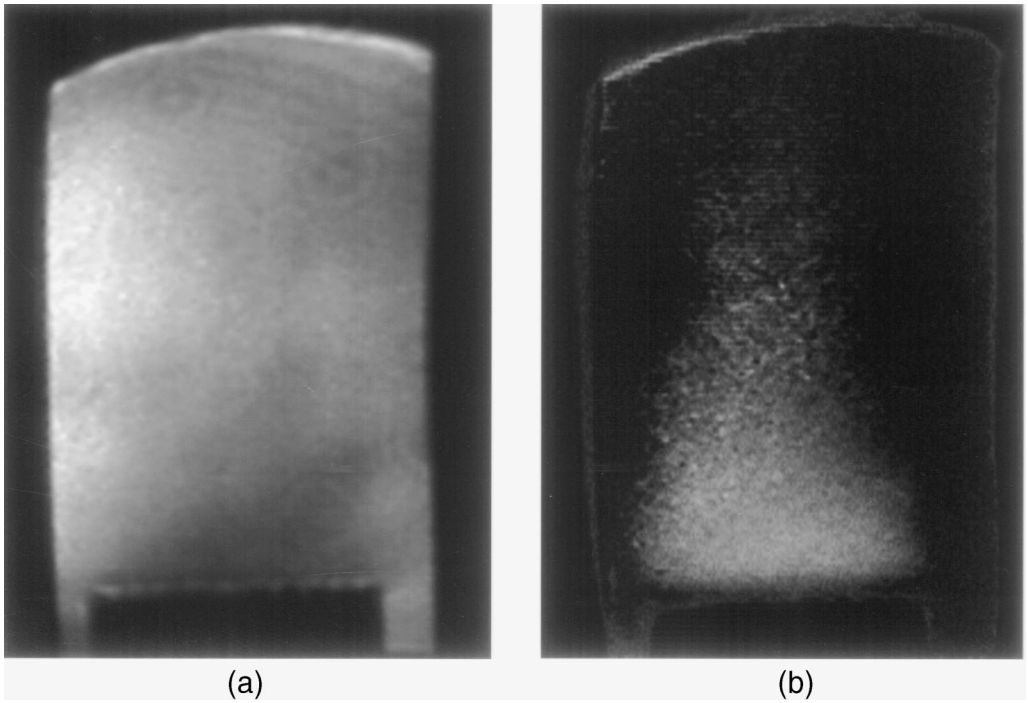

Fig. 2. (a) Probe beam background plus image. (b) Demodulated image extracted from (a).

to the autogaining nature of this borrowed camera (the autogain loop was too slow to affect demodulation). We know, however, that the average concentration of K represented by the image is just under 10^{12} cm^{-3}, which is nearly optically thin. One can, in fact, see that there is no probe absorption visible in Fig. 2(a). It is not possible to detect lower concentrations than this with the current system because an 8-bit digitizer is incapable of resolving better than ~1% modulation in a large background. The edges visible in Fig. 2(b) are the edges of the mirror used to direct the image onto the screen, and they contain some scattered modulated pump light (which can be eliminated by spatial filtering). Finally, we independently detuned the laser and then aspirated pure water through the burner so as to observe no image, thereby ensuring that we were indeed imaging K.

The entire system was devised to accommodate the slow optical modulation speed of the liquid crystals. If we were using electro-optics, for example, we could demodulate the probe images at frequencies in the megahertz regime, which would permit the job to be accomplished within one camera frame. The background problem remains, and it imposes a high detection limit because an 8-bit system can resolve a modulation depth of only 1%. To overcome this problem in pump–probe spectroscopy, one would need to acquire the dc signals simultaneously (in a second detector array) and subtract the two images as analog signals before digitizing. Alternatively, one could use polarization spectroscopy[5] or antiresonant-ring transient spectroscopy.[6] These are crossed-beam, background-free techniques. The low signals produced might require the use of an image intensifier as the demodulator instead of a polarization technique, and this would introduce some reduction in spatial resolution as a trade-off. Alternatively, other conventional rf techniques could be used to enhance the signal-to-noise ratio further.

In summary, we have demonstrated a two-dimensional imaging technique based on pump–probe spectroscopy. It permits high data collection rates, offering the possibility for movies at camera framing rates, and it provides a quenching-independent signal without the need for calibration because each pixel senses an individual modulation depth, independent of individual pixel responsivity or beam profile. In addition, the signal is embedded in a coherent laser beam, making it possible to perform optical processing (spatial filtering is just one example) before the camera.

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